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13. ABSTRACT (Maximum 200 words)

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Final Report on "Reactivity of Highly Excited Diatomic Ions in Hypersonic Nonequilibrium Flow Conditions"

Cheuk-Yiu Ng^{a)} and Rainer A. Dressler^{b)}

a) Ames Laboratory, USDOE and Department of Chemistry, Iowa State University, Ames, IA 50011

b) Air Force Research Laboratory, Hanscom AFB, MA 01731

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During this funding cycle, we have constructed and successfully integrated a state-of-the-art guided-ion beam mass spectrometer into the high-resolution photoion-photoelectron apparatus (Endstation 2) of the Chemical Dynamics Beamline at the Advanced Light Source (ALS) for the study of ion-molecule reaction dynamics. Preliminary tests on the performance of this guided-ion beam mass spectrometer have been made, indicating that the ion-molecule reaction photoionization apparatus is performing well. We have also obtained preliminary results on the collision-induced dissociation (CID) and charge transfer (CT) cross sections for the reactions of $O_2^+(v^+=0-3) + Ar$. Results of these preliminary experiments are briefly discussed below.

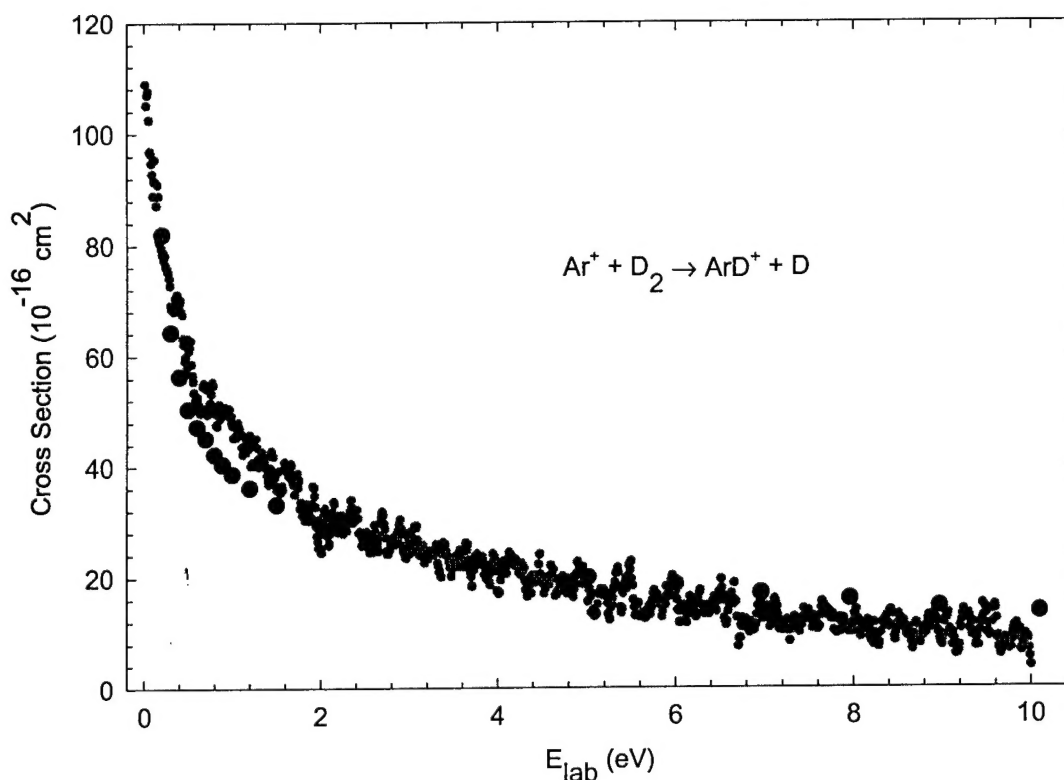
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impact ionization source, we have also examined the CID and CT channels of the $\text{O}_2^+(X^2\Pi_{3/2,1/2g}; v^+) + \text{Ar (Ne)}$ reactions.

II. RESULTS OF PRELIMINARY EXPERIMENTS

A. ArD^+ from $\text{Ar}^+(^2P_{3/2,1/2}) + \text{D}_2$

Figure 1. Absolute total cross sections for reaction (1). The data points in blue are reported by Ervin and Armentrout, whereas those in purple are obtained using the ion-molecule reaction photoionization apparatus at the ALS.

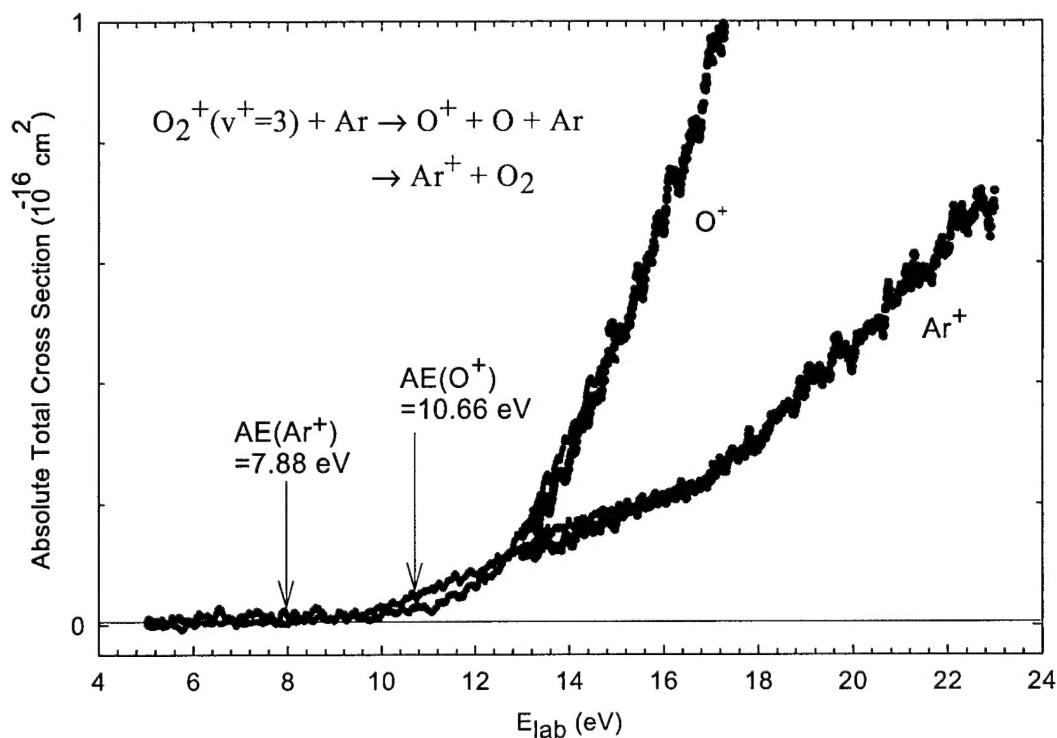


Absolute total cross sections for reaction (1) have been accurately measured by Ervin and Armentrout [*J. Chem. Phys.* **83**, 166 (1985)] over a wide collision energy range. These cross-sections for the formation of ArD^+ (blue dots) in the laboratory kinetic energy (E_{lab}) range of 0.2-10 eV are compared in Fig. 1 to those (pink dots) measured in the E_{lab} range of 0.02-10 eV using the newly constructed ion-molecule reaction photoionization apparatus at the ALS.



As shown in Fig. 1, the general agreement between the two experiments are good after taking into account uncertainties of both experiments. The reactant $\text{Ar}^+(^2\text{P}_{3/2,1/2})$ ions are prepared by electron-impact ionization in the experiment of Ervin and Armentrout, while those in the ALS experiment are formed by photoionization. The good agreement observed in this test experiment indicates that the kinetic energy resolution of the ion-molecule reaction photoionization apparatus is capable of measuring reaction cross sections down to thermal collision energies. **O^+ from $\text{O}_2^+(\text{X}^2\Pi_g, v^+=3) + \text{Ar}$**

Figure 2. Absolute total cross sections for the formation of O^+ and Ar^+ from $\text{O}_2^+(v^+=3) + \text{Ar}$ in the E_{lab} range of 5-23 eV.



In a recent experiment, we have measured the CID and CT cross sections for the reaction of $\text{O}_2^+ + \text{Ar}$ (Ne) [Dressler et al., J. Chem. Phys. **113**, 8561 (2000)]. In this experiment, reactant O_2^+ ions are prepared by ionization of O_2 using an electron beam at $13.0 \pm 0.5 \text{ eV}$. Thus, O_2^+ can be formed in a distribution of vibrational states with v^+ up to 6.

Figure 2 shows the absolute total cross sections for O^+ and Ar^+ formed by reactions (2) and (3), respectively, obtained using the ion-molecule reaction photoionization apparatus.



In this experiment, reactant O_2^+ ions are predominantly formed in the $v^+=3$ state using the nearby resonance autoionizing Rydberg level just above the IE for the formation of $O_2^+(v^+=3)$. As expected, although these cross section curves are similar to those reported recently by Dressler et al., the absolute cross sections are different. The thresholds or appearance energies (AEs) observed for O^+ and Ar^+ (marked in Fig. 2) are in excellent agreement with the thermochemical thresholds of 10.66 eV and 7.88 eV for reactions (2) and (3), respectively. The step observed for the Ar^+ cross-section curve in the E_{lab} range of 12-18 eV is indicative of the opening of the CID channel. This experiment demonstrates that reaction cross sections down to the 10^{-19} cm^2 can be measured using the ion-molecule reaction photoionization apparatus at the ALS.

III. PFI-PESICO MEASUREMENTS

The ultimate goal of this proposed experimental project is to perform PFI-PESICO measurements on state-selected diatomic ions. Instead of detecting the correlated PFI-PEs and PFI-PIs formed in the PFI process, we would look for correlated PFI-PEs and secondary ions formed by the reaction of PFI-PIs. The PFI-PESICO experiments are significantly more difficult than PFI-PEPICO experiments [Jarvis et al., *Rev. Sci. Instrum.* **70**, 3892 (1999)] because the intensities for reaction product ions resulting from collisions between the PFI-PIs and neutral reactants are usually less than 1% that of the PFI-PIs.

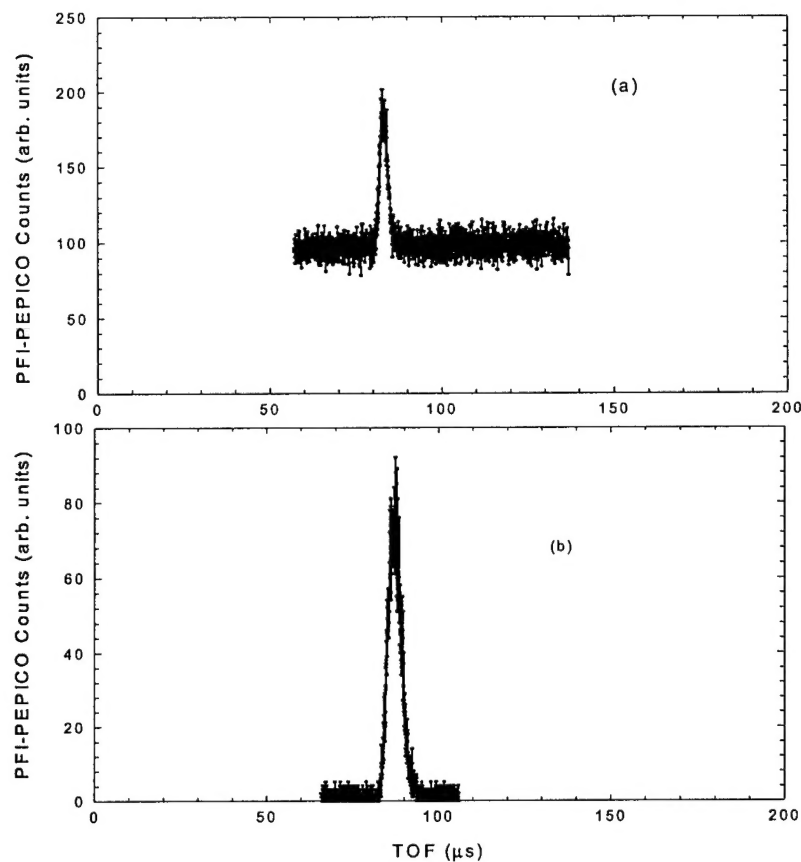


Figure 3. PFI-PEPICO bands for $\text{Ar}^+(^2P_{3/2})$ obtained (a) without using the wired gate and (b) using the wired gate.

Furthermore, the product ions are most likely scattered and dispersed in time, arriving at the ion detector with a significantly broader temporal distribution. Thus, the false coincidence background is expected to be much higher than that observed in PFI-PEPICO measurements for unimolecular dissociation studies. The ion-molecule photoionization apparatus employs two octopole ion-guides for confining reactant and product ions and a QMS for mass detection of reactant and product ions. It is well known that the rf-field is a non-conservative field and thus rf-devices, such as the QMS and octopoles, have the effect of broadening the ion TOF peaks. The PFI-PEPICO TOF peak for Ar^+ observed previously using a linear TOF mass spectrometer was found to have a

FWHM of ≈ 300 ns. This compares to FWHMs of 3-6 μ s for the $\text{Ar}^+(^2\text{P}_{3/2})$ PFI-PEPICO TOF peaks [see Figs. 3(a) and 3(b)] observed using the ion-molecule reaction photoionization apparatus. Consequently the false coincidence background for the PFI-PEPICO band for $\text{Ar}^+(^2\text{P}_{3/2})$ found in Fig. 3(a) is expected to be 10-20 times higher than that observed using the linear TOF mass spectrometer. We note that the PFI-PEPICO peak for $\text{Ar}^+(^2\text{P}_{3/2})$ depicted in Fig. 3(a) was obtained using procedures similar to those reported previously. The false coincidence background of the spectrum shown in Fig. 3(a) has the same intensity as that for the coincidence Ar^+ peak. This high false coincidence background may prevent successful PFI-PESICO measurements. We have overcome this difficulty by using a novel wired gate lens [Brock et al., Rev. Sci. Instrum. **71**, 1306 (2000)] to eliminate most of the false coincidence ions. Using this lens, no ions are allowed to transmit to the ion detector unless a PFI-PE is detected at the electron MCP detector, which signifies the creation of a "true" coincidence ion. The wired gate lens is basically a grid lens constructed by parallel wires with a separation of 0.0125" between adjacent wires. By applying alternative positive and negative potentials of identical magnitudes to these wires, ions are effectively deflected away from the beam path as they pass through the grid. Because of the small distance between adjacent wires, a small potential difference of a few volts applied to adjacent wires is sufficient for deflecting the false ions. This deflecting field should not affect the trajectories of ions at a finite distance from the wired gate lens because the alternate positive and negative potentials seen by these ions are essentially canceled. As shown in Fig. 3(b), the PFI-PEPICO TOF peak for $\text{Ar}^+(^2\text{P}_{3/2})$ obtained using the wired gate lens has nearly zero false coincidence background. This is a very encouraging result. After fine-tuning the operation conditions of the wired gate lens, we would be ready to perform state-selected ion-molecule reaction studies using the ion-molecule reaction photoionization apparatus.

IV. REFEREED JOURNAL PUBLICATIONS

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Final Report on "Reactivity of Highly Excited Diatomic Ions in Hypersonic{PRIVATE} Nonequilibrium Flow Conditions"

Cheuk-Yiu Ng^{a)} and Rainer A. Dressler^{b)}

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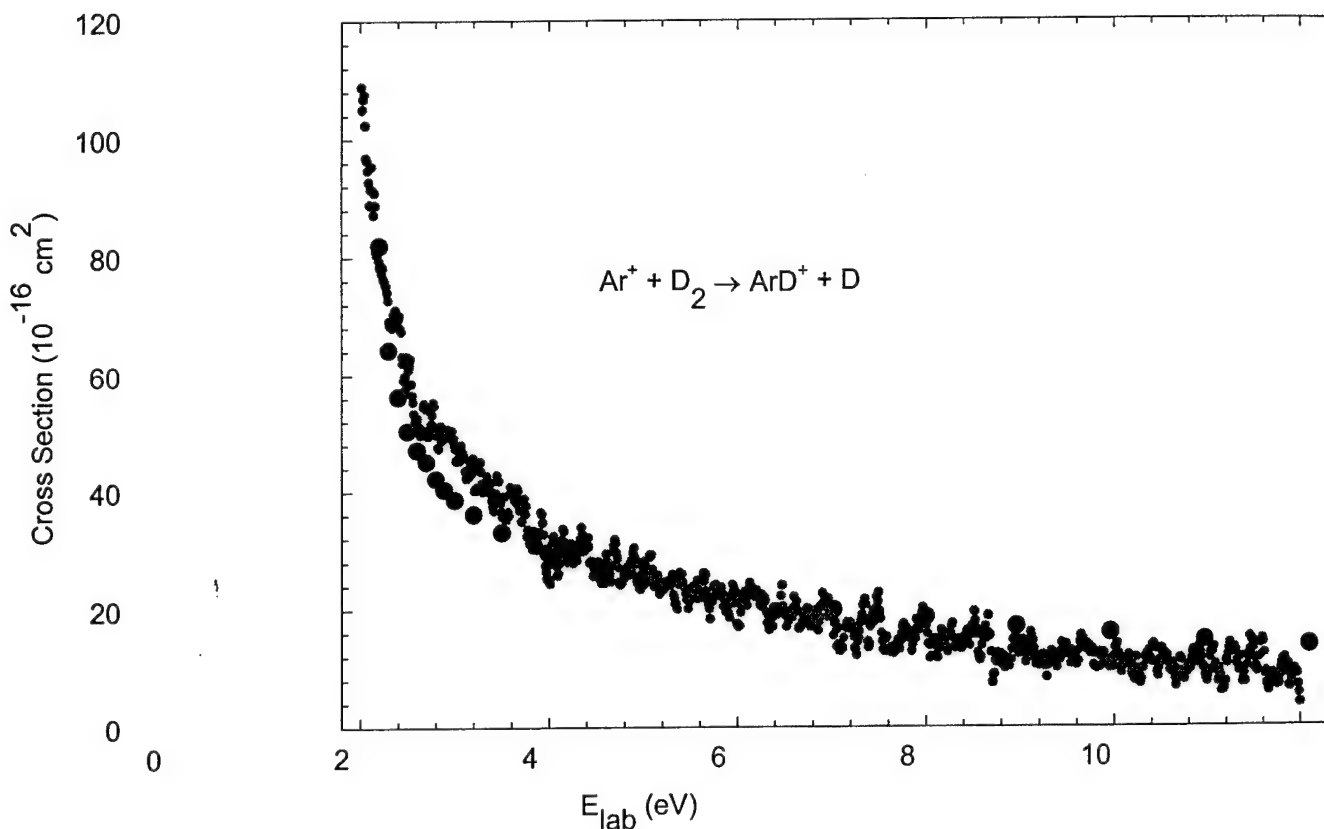
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II. RESULTS OF PRELIMINARY EXPERIMENTS

A. ArD^+ from $Ar^+(^2P_{3/2,1/2}) + D_2$

Figure 1. Absolute total cross sections for reaction (1). The data points in blue are reported by Erwin and Armentrout, whereas those in purple are obtained using the ion-molecule reaction photoionization apparatus at the ALS.

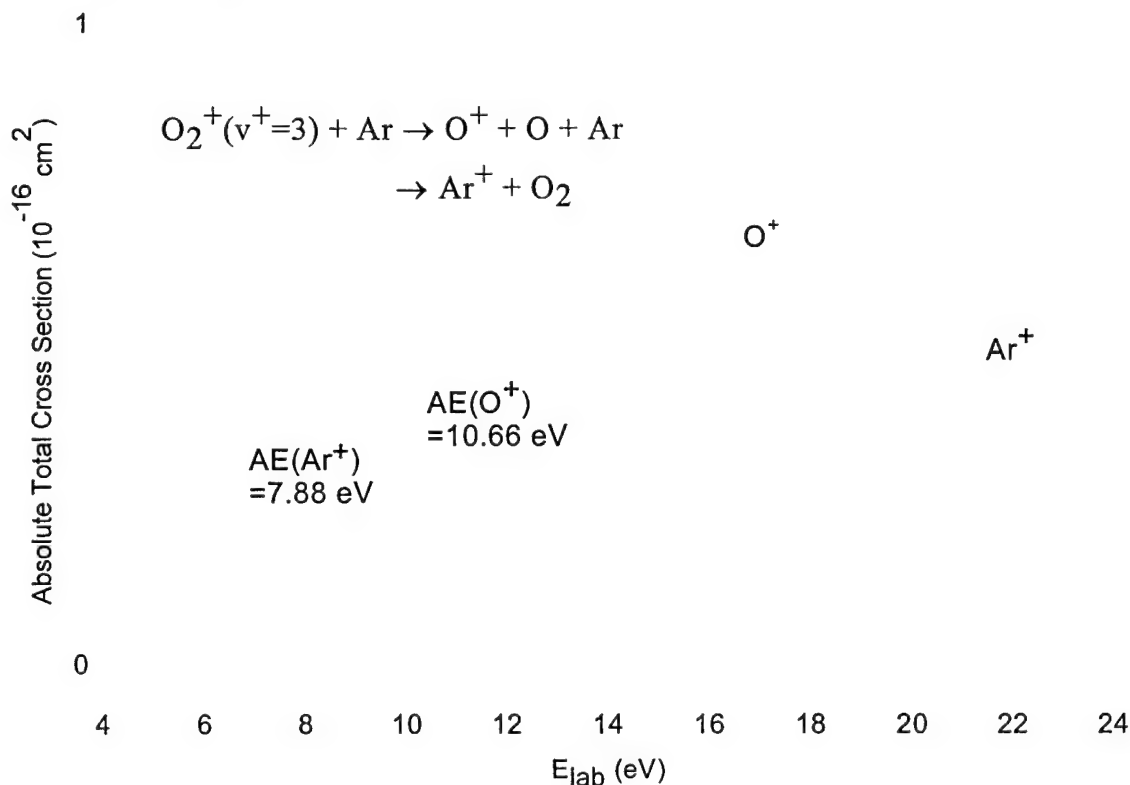


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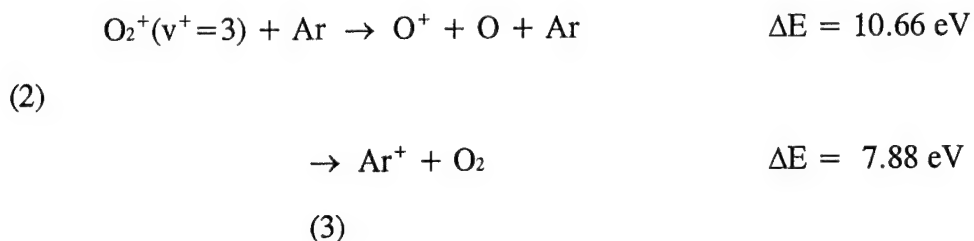
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Figure 2. Absolute total cross sections for the formation of O^+ and Ar^+ from $\text{O}_2^+(v^+=3) + \text{Ar}$ in the E_{lab} range of 5-23 eV.



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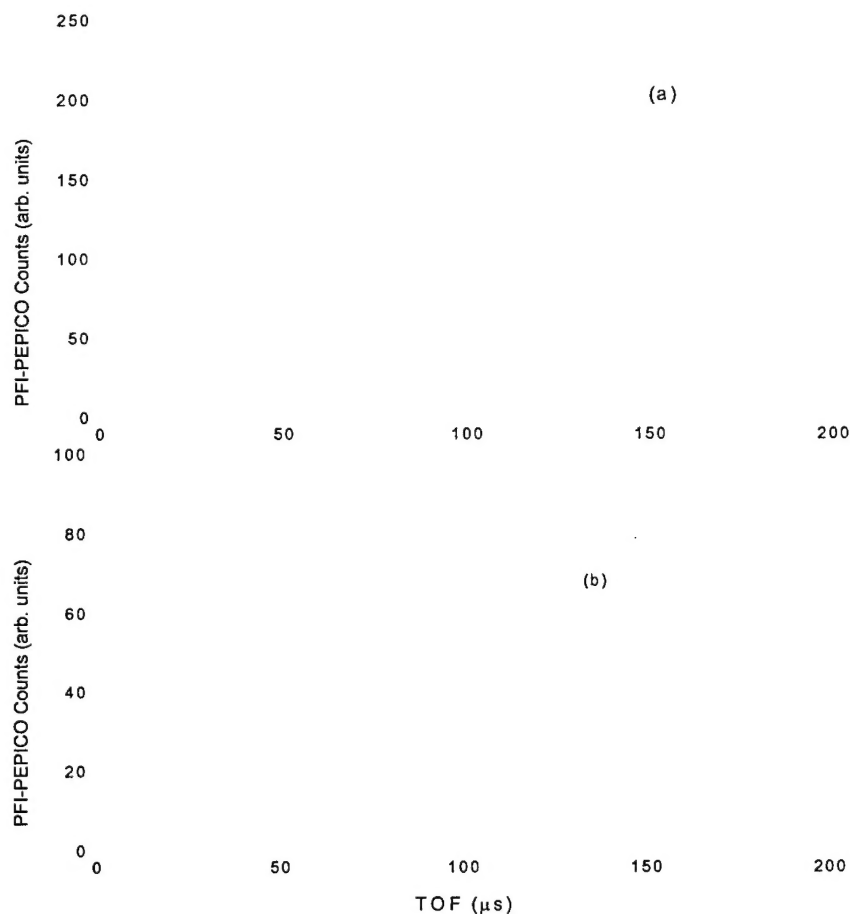


Figure 3. PFI-PEPICO bands for $\text{Ar}^+(^2P_{3/2})$ obtained (a) without using the wired gate and (b) using the wired gate.

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